

# PATENT SPECIFICATION

NO DRAWINGS

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## COMPLETE SPECIFICATION

### Post-Formable Polyethylene Terephthalate Film

We, E. I. DU PONT DE NEMOURS AND COMPANY, a Corporation organised and existing under the laws of the State of Delaware, United States of America, of Wilmington 98, Delaware, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to synthetic organic polymeric film and more particularly to post-formable polyethylene terephthalate film and to its production.

The production of film and fibre forming polyesters of terephthalic acid and a glycol of the series  $\text{HO}(\text{CH}_2)_n\text{OH}$  where "n" is an integer from 2 to 10, inclusive, is disclosed in Specification No. 578,079. From a commercial standpoint, one of the most attractive polymers of this class is polyethylene terephthalate. Polyethylene terephthalate film, when highly oriented and fully heat-treated, i.e. film stretched by substantially the same amount in both directions (e.g. at least 3X, where X is the original dimension of the film) and heat-treated (heat-set) at elevated temperatures (e.g. 150°—220°C.), possesses a combination of electrical, chemical, and physical properties which make it outstanding for a great variety of commercial end uses. However, commercially available polyethylene terephthalate film has not found widespread acceptance in applications which require the film to be post formed into shaped articles of manufacture such as flexible packages to preserve perishable foods because an excessive number of film failures (ruptures) occur in the conventional vacuum forming operation.

According to one aspect of the invention film of substantially amorphous polyethylene terephthalate is stretched at a temperature of 85° to 96°C. in the longitudinal direction (i.e. direction of extrusion or machine direction) by

2.7 to 2.9 times its original dimension in this direction (2.7 to 2.9X), and at a temperature of 85° to 90°C. in the transverse direction by 2.7 to 2.9X; and thereafter heat-set at a temperature of 155° to 165°C. while held under tension. Films so obtained have the following characteristics, which have been found to be critical for the present purpose:

1. a density of 1.3781 to 1.3835;
2. a longitudinal direction  $F_{50}$  value (as defined below) of 13,000 to 18,000 p.s.i.;
3. a transverse direction  $F_{50}$  value no greater than 27,000 p.s.i.;
4. a shrinkage as measured in the transverse direction no greater than 1.5% in water at 100°C.;
5. a refractive index as measured in the longitudinal direction of 1.62—1.64; and
6. a refractive index as measured in the transverse direction of 1.61—1.655.

The invention is predicated primarily upon the discovery that satisfactory post-formability of oriented polyethylene terephthalate film results when amorphous polyethylene terephthalate film is biaxially oriented and heat-treated under conditions such that the interdependent characteristics of  $F_{50}$  values, density, refractive index and shrinkage of the oriented, heat-treated film are within the critical ranges defined above. Films having these characteristics are another aspect of the invention.

The  $F_{50}$  value is defined as the force in p.s.i. (pounds per square inch) calculated from a stress-elongation curve at an elongation of eighty percent. It is measured at 74°C. and 45—50 percent relative humidity. Film samples are cut to 3" x 5" using a cutting die. The film is then further cut to 1" x 5" using a Thwing-Albert cutter. This is done to ensure nick free edges and thus minimize premature breakage. The thickness of the film

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sample is then determined by measurement with an Ames gauge or by weighing and calculation from the density. The sample is then placed in an Instron Tensile Tester. The initial jaw separation is 2 inches. The film is elongated at a constant rate of 2 in./min: (100%/min.) and the force is recorded on a strip chart which is also run at 2 in./min. Elongation is continued until the film fractures. The resultant curve is a force elongation curve. From this curve, break tensile, break elongation and  $F_{60}$  are determined with the tensile values calculated on the basis of the initial cross sectional area. The break tensile is the force corresponding to the point where the film fractured. The break elongation is the elongation at fracture. The  $F_{60}$  is the force at 80 percent elongation (based on original length). Under the conditions of the test, this corresponds to 1.6 inches along the elongation axis.

With respect to the first named characteristic it has been determined that the  $F_{60}$  value measured in the longitudinal direction of the film, hereinafter referred to as the  $LDF_{60}$  value, is a critical parameter in defining a post-formable polyethylene terephthalate film, and must be within the range of from 13,000 to 18,000 pounds per square inch (p.s.i.). To obtain a film having a  $LDF_{60}$  value within the range of from 13,000 to 18,000 p.s.i., it is necessary to stretch the film in the longitudinal direction within a critically narrow range. If the film is stretched in the LD (longitudinal direction) to an extent beyond 2.9X, too high an orientation level will be reached with the result that the  $F_{60}$  value will be in excess of 18,000 p.s.i. and the formability will be seriously impaired by reason of excessive embrittlement of the film during the forming operation. If the film is stretched to an extent less than 2.7X, too low an orientation level will be attained with the result that the  $LDF_{60}$  value will be less than 13,000 p.s.i. and the operability of the process and the physical properties of the film will be inferior. In addition the probability of gel particles being prominent in the stretched sheet and unusually high variations in thickness (gauge) occurring will be high. Stretching the film in the transverse direction of the film to the same extent will also insure  $F_{60}$  values in the transverse direction within the limits essential to good formability.

The operable temperature ranges employed for LD stretching the film will depend upon the method used for longitudinal direction stretching. If an idler roll system (such as is shown, for example, in Specification No. 743,503) is employed, the operable LD stretching temperature range will be 90—96°C. Film heated above 96°C. will stick to the rolls and wrap around them causing non-uniform stretching. Film heated below 90°C. will neck-in excessively, edge-heavying will occur,

and the film surface will be more prone to scratches. If a nip-roll type stretching device is used, the operable LD stretching temperature range will be 85—92°C. Above 92°C., the film will again stick to the roll creating lanes of irregular stretch. Below 85°C., the film will not stretch uniformly at the rates normally employed for stretching.

The film may be stretched to the required extent by stretching the film first in the transverse direction in a tentering device followed by longitudinal direction stretching in a nip-roll stretching apparatus such as described in Specification No. 890,004, or by stretching film first in longitudinal direction in an idler roll apparatus such as described in Specification No. 743,503 and thereafter stretching the film in the transverse direction in a tentering device similar to that described.

At the temperatures employed for stretching in this process the rates of stretch may range from 5000 percent per minute for thick gauge films, to 65,000 percent per minute for thin gauge films.

Density, which is a measure of the level of crystallinity of the film, is also a critical film characteristic. Density values ranging from 1.3781 to 1.3835 have been found to be acceptable. Density levels in excess of 1.3835 lead again to reduced formability levels. Densities below 1.3781 lead to poor dimensional stability (excessive shrinkage at elevated temperatures) which seriously affects the productivity of desired post-treatments such as coating. The final density of the film is determined by the heat-treating step. If the film is heat-treated above a temperature of 165°C., the density level, and hence the level of crystallinity, will be too high. Below 155°C., poor dimensional stability and excess shrinkage will be the result.

The refractive index of the film is a direct measure of orientation, and together with density is correlated with the  $F_{60}$  value. A film having an  $LDF_{60}$  value between 13,000 and 18,000 and a density between 1.3781 and 1.3835 will have an LD refractive index between 1.61 and 1.64. The  $TDF_{60}$  is less critical but must be between 12,000 and 27,000 p.s.i. Films of the proper density and  $TDF_{60}$  range will have TD refractive indices between 1.61 and 1.655.

The process of the present invention, although, as illustrated by the examples to follow, it is particularly effective in producing oriented polyethylene terephthalate films possessing excellent post-forming characteristics wherein the thickness of the stretched film lies between 0.5—2.0 mils, is applicable to polyethylene terephthalate films 0.25—10 mils thick. For some end uses, wherein it may be desirable to employ films of thicknesses greater than 10 mils, it is possible to prepare such films with good post-forming characteristics by compensating for the differ-

ences in rates of crystallization and requisite stretching forces in these thick gauge films by means of proper alteration of the process conditions.

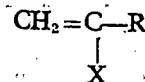
- 5 For many end uses of the thermo-formable film, such as in the manufacture of flexible packages to protect perishable foods, it is not only desirable, but at times necessary, to coat the base of polyethylene terephthalate film with an oxygen and moisture impermeable coating. In this connection the extent of transverse direction shrinkage at 100°C. in water is a significant parameter to insure good coating performance of the base film. Maximum TD shrinkage of 2.5% is acceptable. Experimental data show that such a film can be coated at speeds in excess of 125 yds. per minute. TD shrinkage (measured at 100°C.) below 1.5% is preferred since this enables the film to be coated at speeds of 170 yds. per minute or greater. A transverse direction stretch ratio between 2.7 to 2.9X, in combination with the above-specified density requirements, produces a film with satisfactory shrinkage.

The base film of the process of the present invention may have formed thereupon:

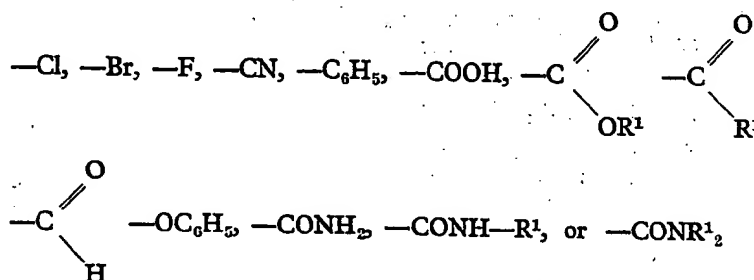
1. a moisture and oxygen impermeable coating, and/or,
2. a heat-sealable coating readily adaptable to surface modifications as printing etc.

As suitable moisture and oxygen impermeable coating compositions may be listed solid polymers prepared from at least 80% vinylidene chloride and 3 to 20% of at least one

other polymerizable mono-olefinic monomer. As polymerization monomers for use with the vinylidene chloride may be listed, methyl, ethyl, isobutyl, octyl and 2-ethyl hexyl acrylates and methacrylates: phenyl methacrylate, cyclohexyl methacrylate, p-cyclohexylphenyl methacrylate, methoxy ethyl methacrylate, chloroethyl methacrylate, 2-nitro-2-methyl propylmethacrylate, and the corresponding esters of an acrylic acid: methyl- $\alpha$ -chloroacrylate, octylalphi-chloroacrylate, methylisopropenyl ketone, acrylonitrile, methacrylonitrile, methylvinyl ketone, vinyl chloride, vinyl acetate, vinyl propionate, vinylchloroacetate, vinyl bromide, styrene, vinyl anaphthalene, ethyl vinyl ether, N-vinylphthalimide, N-vinylsuccinimide, N-vinylcarbazole, isopropenyl acetate, acrylamide, methacrylamide or monoalkyl substitution products thereof, phenyl vinyl ketone, diethyl fumarate, diethylmaleate, methylene diethyl malonate, dichlorovinylidene fluoride, dimethyl itaconate, dibutyl itaconate, vinyl pyridine, maleic anhydride, allyl glycidyl ether and other unsaturated aliphatic ethers described in U.S. Patent No. 2,160,943. These compounds may be described as vinyl or vinylidene compounds having a single  $\text{CH}_2=\text{C}<$  group. The most useful ones fall within the general formula



where R is hydrogen, halogen or a saturated aliphatic radical, and X is



in which  $\text{R}^1$  is alkyl.

- 70 Polymeric coating compositions which can be employed as heat-sealable coatings for the post-formable polyethylene terephthalate film of the present invention are the polymers, e.g., homopolymers and copolymers, of alpha-olefins, such as polyethylene and copolymers of propylene and butylene, etc.

The coatings may be applied from aqueous or organic vehicles, i.e., in the form of aqueous dispersions or from solutions of the polymers in organic solvents, and may be applied in

accordance with any known coating techniques. They may be applied, for example, by passing the film through a bath in a continuous manner or in a batch manner. Coatings may also be sprayed on the film, or applied manually by brushing or the like. The thickness of the coatings may be adjusted in accordance with methods well known in the coatings art. Selection of the proper thicknesses of the coating depends to some extent on the properties that one desires to emphasize.

The following examples will serve to further illustrate the principles and practice of this invention.

EXAMPLE 1.

5 Molten polyethylene terephthalate having an intrinsic viscosity of 0.54 was cast from a pressurized extrusion apparatus onto a water cooled quench drum at a throughput of 900 to 1000 pounds per hour. The temperature of the molten polymer was approximately 280°C. The extruded film was quenched by contact with the quench drum which was maintained at 40°C. The resultant cast film was approximately 0.04 inch thick and approximately 40 inches wide. This cast film was heated to a temperature of 94°C. and was then stretched in the longitudinal direction over an idler roll stretching apparatus such as is described in U.S. Patent 2,823,421 to an extent of 2.9X. After LD stretching, the film was heated to 89°C. and was then stretched to an extent of 2.9X in a tentering device such as described in Specification 743,503. Heat-treating of the film was accomplished by the heating of the film to a temperature of 161°C. while holding the film under tension in an extension of the tentering device. The resulting film had an  $LDF_{50}$  value of 16,500 p.s.i., a density of 1.3817, an LD refractive index of 1.634, and a TD shrinkage in water at 100°C. of 1.4%. After heat-treating, the film was coated on one side with a copolymer comprising 90% vinylidene chloride/10% of acrylonitrile and 1% based on the weight of vinylidene chloride and acrylonitrile of itaconic acid to confer impermeability to water vapor and oxygen. The resulting film was then subsequently top coated with a 3-mil coating of polyethylene by conventional melt extrusion methods. The coated film was converted into flexible packages on a conventional vacuum and heat package forming machine. The coated film exhibited excellent formability and adaptability to this type of standard vacuum pack-

aging apparatus, exhibiting superior resistance to excessive shrinkage and rupture ("blow-out").

EXAMPLES 2 TO 12.

These examples illustrate the criticality of the  $LDF_{50}$  value, density, and percent of TD shrinkage as measured at 100°C., in the preparation of post-formed polyethylene terephthalate film. In a manner identical to that described in Example 1, molten polyethylene terephthalate having an intrinsic viscosity of 0.54 was cast from a pressurized extrusion apparatus onto a water cooled quench drum and was quenched by contact with the quench drum. Samples of the cast film were stretched in the LD in a manner identical to that described in Example 1 to varying extents between 2.6 to 3.3X. After LD stretching, the films were stretched in a tentering device as in Example 1 to extents varying between 2.8 and 3.0X. The films having a thickness of 0.5 mils were then heat-treated in the extension of the tentering device while being held under tension at a temperature between 125 to 165°C. The heat-treated film samples were tested for such physical properties as  $LDF_{50}$ , density, dimensional stability at 100°C. in water, and 150°C. in air prior to being coated on one side with a 4.0 gram per square meter copolymeric coating composition comprising 90% vinylidene chloride, 10% acrylonitrile and 1% of itaconic acid (based on the weight of vinylidene chloride and acrylonitrile) such as is described in Example 1. The physical properties data for these films, the process conditions, the maximum permissible speed at which the film could be coated (without causing wrinkling in the coating apparatus and poor roll formation), and the post formability of the coated film on standard vacuum and heat package forming machinery after it had been top-coated with a 3-mil thickness of polyethylene as described in Example 1, are listed in Table I.

TABLE I  
Process Conditions and Physical Properties of Post-Formable Polyethylene Terephthalate Film

Example No.	Stretch Ratio		Stretch Temperature °C.		Heat-Treating Temperature °C.	LD F-80 p.s.i.		TD F-80 p.s.i.	Density g/cc.	Refractive Index	
	LD	TD	LD	TD		LD	TD				
2	2.8	2.8	94	89	156	16,900	19,800	1.3826	1.6330	1.6510	
3	2.7	2.8	94	89	156	13,300	24,200	1.3800	1.6261	1.6540	
4	2.9	2.8	94	89	155	17,300	19,300	1.3812	1.6339	1.6472	
5	2.9	2.9	94	89	165	16,500	—	1.3817	1.6340	1.6490	
6	3.3	3.0	94	89	125	23,900	23,700	1.3690	1.6307	—	
7	3.3	3.0	94	89	145	23,900	25,200	1.3725	1.6385	1.6479	
8	3.3	3.0	94	89	155	23,900	—	1.3816	1.6406	1.6530	
9	2.6	2.8	94	89	155	12,700	19,800	1.3810	1.6205	—	
10	2.7	2.8	94	89	155	13,800	20,600	1.3810	1.6220	—	
11	3.0	2.9	94	89	155	22,000	—	1.3810	1.6390	—	
12	2.8	2.9	94	89	180	19,600	—	1.3852	—	—	

TABLE I (Continued)

Example No.	Dimensional Stability in water at 100° C.		Dimensional Stability in air at 150° C.		Maximum Permissible Coating Speed yd/min.	Formability Heat on vacuum Heat Packaging Machine
	LD	TD	LD	TD		
2	0.8	1.25	7	7	—	Good
3	1.6	0.8	8.7	5.4	155 — 160	Good
4	1.4	1.4	7.9	6.7	—	Good
5	1.5	1.4	6.5	6.0	140	Good
6	8.8	11	16.8	19.3	60 — 80	Poor
7	3.8	5	—	—	65 — 90	Poor
8	3.2	4.0	14.1	15.9	65 — 90	Poor
9	—	—	—	—	—	Poor
10	—	—	—	—	—	Good
11	—	—	—	—	—	Poor
12	—	—	—	—	—	Poor

Example 1, samples of polyethylene terephthalate were prepared in accordance with the process specifications of the present invention as stated hereinbefore. Representative samples of these films were tested for such physical properties as tensile modulus, tensile strength, percent elongation and dimensional stability at 100°C. in water. From the data in Table II it can be seen that the post-formable polyethylene terephthalate film prepared by the process of the present invention possesses physical property levels well within the physical specifications set forth for commercially available polyethylene terephthalate film.

LDF<sub>50</sub> values far in excess of 18,000 p.s.i., the ability to be converted into flexible formed packages by the conventional vacuum packaging machinery was seriously impaired. In Examples 6 and 7, the heat-treating temperatures were without the specified limits and as a consequence a TD shrinkage at 100°C. in water far in excess of the 2.5% limit was obtained. It was therefore impossible to coat the film at economically feasible speeds. In Example 12, the heat-treating temperature was also in excess of 165°C. and as a consequence, formability on the conventional vacuum packaging machinery was poor.

EXAMPLES 13 TO 16.

In a manner identical to that described in

As can be seen from Table I, a polyethylene terephthalate film (Examples 2 to 5) processed according to the specifications of the invention set forth hereinabove exhibits all the characteristics necessary to produce a satisfactory post-formable film. In Examples 6-8, 9, 11 and 12, wherein the specifications of the process of the present invention were not adhered to, polyethylene terephthalate possessing poor post-forming characteristics were obtained. By introducing too low a level of orientation (stretching less than 2.7X LD) as evidenced in Example 9 by a LDF<sub>50</sub> value less than 13,000 p.s.i., or too high a level of orientation (stretching in excess of 2.9X LD) as evidenced in Examples 6, 7, 8 and 11 by

TABLE II

Process Conditions and Physical Property Levels of Post-Formable Polyethylene Terephthalate Film

Example No.	Stretch Ratio		Stretch Temperature °C.		Heat Treating Temp. °C.	Modulus 10 <sup>-5</sup> p.s.i.		Tensile Strength 10 <sup>-3</sup> p.s.i.		Elongation Percent		LD Refractive Index	Density g/cc.	Dimensional Stability Percent					
	LD	TD	LD	TD		LD	TD	LD	TD	LD	TD			100 °C./water		150 °C./air			
														LD	TD	LD	TD	LD	TD
13	2.8	2.9	94	89	161	6.25	5.82	.275	.255	.123	125	1.6400	1.3802	1.4	1.5	7.8	8.1		
14	2.8	2.8	94	89	156	7.00	6.55	.249	.202	.110	127	1.6330	1.3826	1.4	1.2	7.5	7.5		
15	2.7	2.7	94	89	156	5.27	7.09	.220	.281	.152	105	—	1.3824	1.6	0.8	5.8	6.0		
16	2.9	2.8	94	89	155	5.26	6.19	.227	.246	.126	125	—	1.3821	1.1	1.1	8.0	7.3		
Control	3.0	3.0	94	89	200—220	6.99	8.0	.251	.281	105.2	124	—	1.3950	0.5	0.75	1.0	1.75		

## EXAMPLES 17 TO 21.

In a manner similar to that described in Example 1, polyethylene terephthalate films (0.5 mils in thickness) were prepared. Examples 17, 20 and 21 represent film prepared within the process limitations of a present invention, i.e., LD stretched between 2.7 to 2.9 and 2.8 to 2.9 in the TD. Examples 15 and 16 represent films stretched in excess of the specified process limitations (3.30 LD and 3.0 TD). All film samples were heat-treated at 161°C. and held under tension. Samples of the films which were:

1. uncoated,
2. coated with a 4 gram per square meter coating composition of 94/6/1 vinylidene

chloride/acrylonitrile/itaconic acid, and  
3. coated as in 2 above with a topcoat of 3-mil thick polyethylene,

were tested on a commercial vacuum-heat-forming packaging machine under machine operating conditions. Table III illustrates the performance (expressed in terms of percent of film failures) of these various film samples. As can be seen from Table III, the performance of the polyethylene terephthalate film prepared by the present process on the commercial vacuum and heat package forming machinery far exceeds that of the films prepared outside the process limitations of the invention.



TABLE III

## Evaluation of Post-Formable Polyethylene Terephthalate Film on Commercial Vacuum-Heat Package Forming Machinery

Example No.	Stretch Ratio		Uncoated Base Film	Base Film Coated with O <sub>2</sub> -H <sub>2</sub> O Vapor Impermeable Coating*	Base Film Subcoated with O <sub>2</sub> -H <sub>2</sub> O Impermeable Coating* and Topcoated with Polyethylene**	
	LD	TD			a. under machine setting #1*** b. under machine #2***	
17	2.8	2.8	44	15.8	0.0	5.0
18	3.3	3.0	100	50 — 100	11.0	Burn Out x
19	3.3	3.0	100	43.5	16.0	Burn Out x
20	2.7	2.8	47.5	26	0.0	11.0
21	2.9	2.9	44	14	0.0	4.2

\* — 94/6/1 — Vinylidene chloride/acrylonitrile/itaconic acid — 4 g/m<sup>2</sup> coating on one side.

\*\* — 3-mil coating of polyethylene on one side.

\*\*\* — The evaluation of the formability of these films was carried out on the vacuum packaging machine under the two sets of conditions (temperature and machine speed) normally employed in the commercial preparation of packages. Machine setting #2 denotes the employment of a higher heat control setting on the machine; hence a more drastic test.

x — Film melted.

# EXAMPLES 22 TO 26.

Molten polyethylene terephthalate having an intrinsic viscosity of 0.54 was cast from a pressurized extrusion apparatus onto a water cooled drum at a throughput of 150 lbs. per hour in a manner identical to that described in Example 1. Samples of the cast film were heated to a temperature of 90°C. and were then stretched in the longitudinal direction over an idler roll stretching apparatus in a manner described in Example 1. The film samples were stretched to varying extents between 2.8—3.3X. After LD stretching, the films were heated to 89°C. and were then transversely stretched to an extent of 2.9X in a tentering device similar to that described hereinbefore. The bi-directionally stretched

film samples (0.5 mil thick) were then heat-treated under tension in an extension of the tenter frame at temperatures ranging between 162—195°C. The density (gms./cc.) LDF<sub>80</sub> (p.s.i.), and percent shrinkage at 100°C. in water were measured for the film samples. Additionally, these samples were tested in an uncoated state on the commercial vacuum and heat package forming machinery described hereinbefore for percent failures (blow-outs). Table IV lists the stretch ratios employed, heat-treating temperatures, density, percent shrinkage at 100°C. in water, and the percent failures of the various film samples on the vacuum and heat package-forming machinery.

From Table IV it can be seen that when

the process limitations of the present invention were adhered to (Example 22) film possessing excellent post-forming characteristics\* was obtained. In Examples 23 (LD stretch ratio 3.3X) and 24 (heat-treating temperature 195°C.) where the process limitations were not adhered to, films having unacceptable post-forming characteristics were produced.

\*—The uncoated base films were tested on commercial vacuum and heat packaging machinery normally employed for the packaging of coated films. Hence a performance of less than 50% failures of the uncoated base film samples is considered excellent.

TABLE IV

Process Conditions and Physical Properties of Post-Formable Polyethylene Terephthalate Film

Example No.	Stretch Ratio		Heat-Treating Temperature °C.	LD F-80 psi (10 <sup>-3</sup> )	Density gm/cc.	Dimensional Stability in water at 100° C.		Performance of Uncoated Base Film on Commercial Vacuum and Heat Packaging Machinery Percent Film Failures
	LD	TD				LD	TD	
22	2.8	2.9	162	18.8	1.3800	1.6	2.1	up to 38
23	3.3	2.9	162	31.3	1.3792	2.8	3.2	100
24	2.8	2.9	195	19.7	1.3922	0.8	0.6	100

The process of the present invention provides a simple efficient, economical method for preparing heat-treated oriented polyethylene terephthalate film which is readily adaptable to post-forming processes. The product not only may be satisfactorily employed on all types of post-forming equipment but also lends itself ideally to pre-surfacing modifications such as coating or metallizing. The development of this product paves the way for extensive use of polyethylene terephthalate films in a wide variety of packaging applications wherein the packaging film must be formed into a shaped article of manufacture by the use of vacuum-heat package forming equipment: a use hitherto not exploited by this type of film, despite its superior physical properties, because of its questionable post-forming characteristics. The polyethylene terephthalate films of the present invention may be employed for a wide variety of end uses. Among these may be listed: fenders for bicycles, tail-light reflectors, blister packages, interlayer for tubeless tires, vacuum formed food packages, containers and pouches, e.g., pouches for luncheon meats, cheese frankfurters etc., portion control cups, containers for jellies, baby foods and soups, frozen foods, skin packaging applications, typewriter ribbons, cook and serve food packages, stiffeners for waist bands of trousers, shirts, foundation garments etc., and collar stays. A particularly useful application for thick gauge post-formable polyethylene terephthalate films lies in the manufacture of pleated and coiled films for use as decorative corrugated structures and as conductive, lightweight, corrosion resistant, leaf, coil, and torsion type springs. These spring devices, which are functional over a wide range of elongation and demonstrate such desirable properties as extendability, retractability and constant tensions find utility in a multitude of commercial applications. For example, in the form of sheet spring, this device may be used in safety devices, door and window closures, room dividers, rollerless window shades, toothed drive shafts, suspension packaging, shock-absorbing straps, and temperature-moisture sensing devices. Other spring types are capable of providing extendable "snap" antennas, compressible weather stripping and retractable wiring and circuiting. Slats for venetian blinds may be made from transparent post-formable polyethylene terephthalate film which has been vacuum metallized.

#### 55 WHAT WE CLAIM IS:—

1. A process for producing post-formable oriented polyethylene terephthalate film, which comprises stretching a substantially amorphous polyethylene terephthalate film at a temperature of 85° to 96°C. in the longitudinal direction by 2.7 to 2.9 times its original dimension in this direction, and also stretching it at a

temperature of 85° to 90°C., in the transverse direction by 2.7 to 2.9 times, and thereafter heat-setting the film at a temperature of 155° to 165°C., while maintaining it under tension. 65

2. Process according to claim 1, wherein the film is stretched in the longitudinal direction at a temperature of 90° to 96°C. in an idler roll system. 70

3. Process according to claim 1, wherein the film is stretched in the longitudinal direction at a temperature of 85° to 92°C. between sets of nip rolls. 75

4. Process for producing post-formable oriented polyethylene terephthalate film according to claim 1 substantially as hereinbefore described. 80

5. A post-formable oriented polyethylene terephthalate film obtained by a process claimed in any one of claims 1 to 4. 85

6. An oriented polyethylene terephthalate film capable of being post-formed into flexible articles of manufacture characterised by having: a density of 1.3781 to 1.3835, an  $F_{80}$  value (as hereinbefore defined) measured in the longitudinal direction of the film of 13,000 to 18,000 pounds per square inch, an  $F_{80}$  value measured in the transverse direction of the film not greater than 27,000 pounds per square inch, a shrinkage measured in the transverse direction not greater than 1.5% in water at 100°C., a refractive index measured in the longitudinal direction of 1.62 to 1.64, and a refractive index measured in the transverse direction of 1.61 to 1.655. 90 95

7. A film according to claim 5 or 6 having on at least one surface a heat-sealable coating consisting essentially of a solid polymer of an alpha-olefine. 100

8. A film according to claim 7, wherein the coating is of polyethylene. 105

9. A film according to any one of claims 5 to 8, having on at least one surface a coating comprising essentially a copolymer containing 80 to 97% by weight, based on the total weight of the copolymer, of vinylidene chloride, and 20 to 3% by weight of at least one other polymerisable mono-olefinic monomer. 110

10. A film according to claim 9, wherein the said coating comprises a copolymer containing 90% by weight of vinylidene chloride, 10% by weight of acrylonitrile, and 1% by weight, based on the total weight of vinylidene chloride and acrylonitrile, of itaconic acid. 105

11. A film according to any one of claims 7 to 10, carrying on at least one surface:

(a) a sub-coating consisting essentially of a copolymer of vinylidene chloride and another polymerisable mono-olefinic monomer, and 120

(b) a heat-sealable top-coating consisting essentially of a solid polymer of an alpha-olefine. 125

12. A film according to claim 6 substantially as hereinbefore described.

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